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Temperature Trends in Coal Char Combustion under Pressurized Oxy-Combustion Conditions for the Determination of Kinetics

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Abstract:

Oxy-fuel combustion technology with carbon capture and storage could significantly reduce global CO_2 emissions, a greenhouse gas. Implementation can be aided by computational fluid dynamics (CFD) simulations, which require an accurate understanding of coal particle kinetics as they go through combustion in a range of environments. To understand the kinetics of pulverized coal char combustion, a heated flow reactor was operated under a wide range of experimental conditions. We varied the environment for combustion by modifying the diluent gas, oxygen concentration, gas flow rate, and temperature of the reactor/reacting gases. Measurements of reacting particle temperatures were made for a sub-bituminous and bituminous coal char, in environments with CO_2 or N_2 as the diluent gas, with 12, 24, and 36 vol-% oxygen concentration, at 50, 80, 100, and 200 standard liters per minute flowing through the reactor, reactor temperatures of 1200, 1400 K, at pressures slightly above atmospheric. The data shows consistent increasing particle temperature with increased oxygen concentration, reactor temperature and higher particle temperatures for N_2 diluent than CO_2. We also see the effects of CO_2 gasification when different ranks of coal are used, and how the reduction in the temperature due to the CO_2 diluent is greater for the coal char that has higher reactivity. Quantitative measurements for temperature are not yet complete due to ongoing calibration of detection systems.

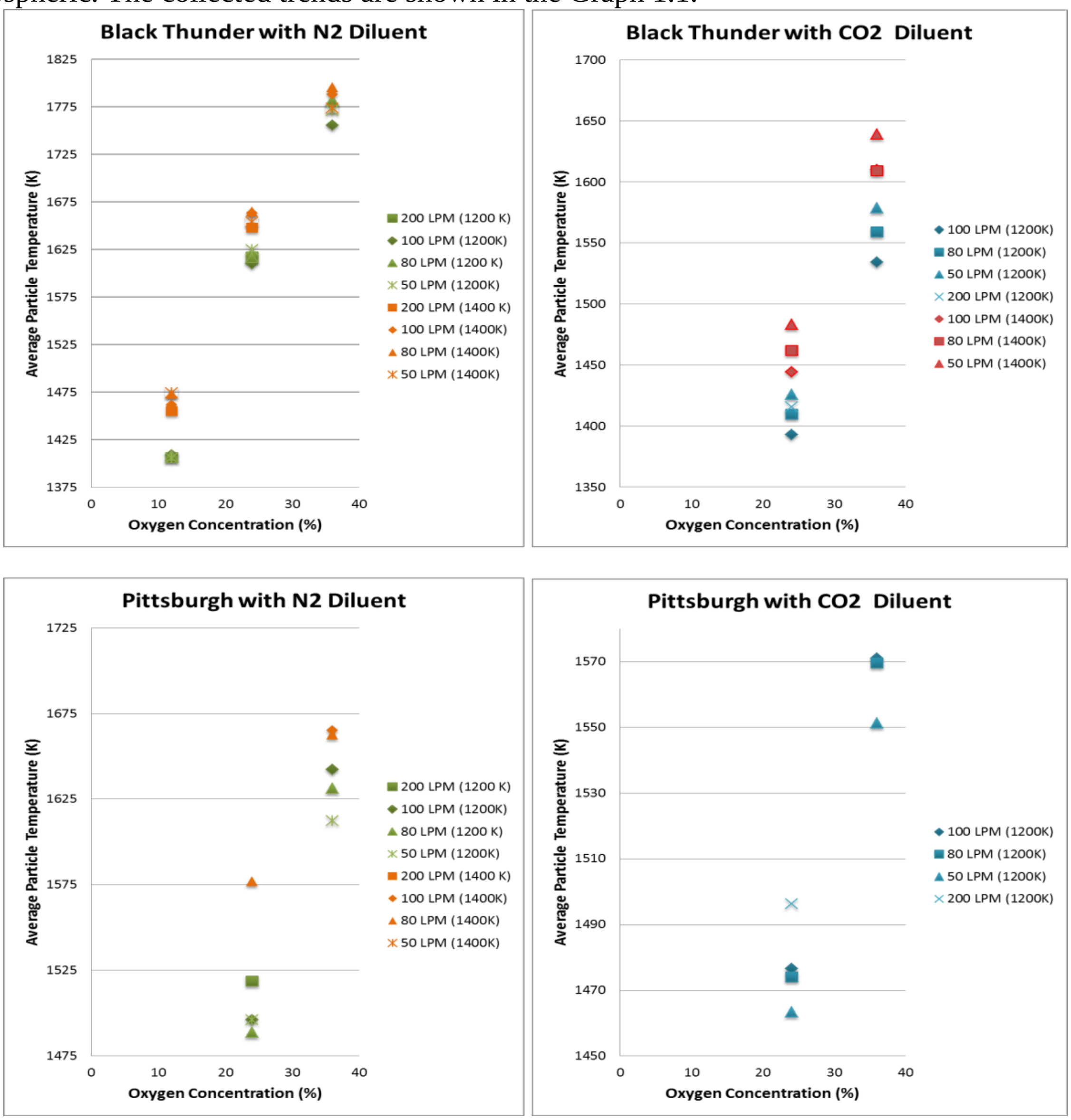
Introduction:

Oxy-fuel combustion of coal has gained attention recently as a promising way to continue to generate electrical power from coal while implementing carbon capture and storage (CCS). This technology can be retrofitted onto existing coal fire power plants, potentially making it more cost effective than building new infrastructure. Utilization of oxy-fuel combustion of coal with CCS could significantly reduce CO_2 emissions and lessen the effects that cause climate change. The efficiency penalty for implementing oxy-fuel combustion with CCS as compared to air fired combustion without any capture systems is around 10 percent, when boilers are operating at 1 atm. When oxy-fuel combustion with CCS occurs at pressure, the efficiency penalty can be reduced to about 7% through better heat integration [1]. Oxy-fuel combustion of coal under pressure also prevents air leakage into the system and may produce lower NO_x emissions. However, in order to effectively implement this technology, there needs to be a deeper understanding of oxy-fuel combustion under pressure. Computational fluid dynamics simulations utilizing kinetic models for pressurized oxy-fuel coal combustion can then be used for system design and process optimization. Our goal is to measure individual coal char particle temperatures as they combust in a pressurized environment, and observe trends under various conditions, to determine these kinetic rates. We varied the environment for combustion by modifying the pressure, diluent gas, oxygen concentration, gas flow rate, and temperature of the reactor/reacting gases. In addition to collecting qualitative temperature data we have the means to also get in-situ measurements of particle size and velocity. Calibration of these detection systems is a work in progress.

Progress:

Approach: To carry out our experiments coal chars are first generated, which are coal particles that have been heated in a 1200 K furnace with nitrogen, releasing moisture and volatile species. These particles are then sieved into narrow size bins, and we used particles with diameters between 90 and 120 μm. The char particles are introduced into a combustion environment and their temperatures are measured. An example of coal char combustion is shown in Figure 1.1. To carry out the combustion reaction we utilize the Pressurized Combustion and Gasification Reactor (PCGR), which is shown in Figure 1.2 along with the schematic. The PCGR is capable of achieving an internal temperature up to 1600°C and 300 psi. The ports on the reactor allow us to insert an optical probe and cold tip to measure light emissions from the reacting particles. The optical probe is shown in Figure 1.3, which collects the light emissions of reacting particles. The probe is inserted into a cooling water sheath that is bolted into the reactor. The cold tip probe is loaded in a port directly across reducing the background thermal radiation seen from the probe so that char particle emissions can be discriminated as the particle falls through the reactor. The probe is connected to a fiber optic bundle that transmits the light to the detectors, and analyzed using two-color pyrometry. The detection system is shown in Figure 1.4 and consists of the pyrometer detectors, control electronics and the data acquisition devices that interface with our LabView programs. The front panel of the optical data acquisition program coded through LabView is shown in Figure 1.5. This program allows us to conduct 2-color pyrometric temperature measurement on individual particles burning in isolation as they fall through the reactor, along with particle size and velocity measurements. The LabView program also shows histograms for measurements on individual particles that pass through the probe location (we get around 100 particles samples per set), and performs real time error checking, real time signal fitting, and can save raw and processed data. Currently we focus on the temperature readings that are calculated using Plank's radiation law assuming that the particles are gray body emitters. There is a calibration constant associated with these equations and the optical system [3]. Particle size or velocity analyses are not fully accurate since there is currently no algorithm to check if the particles are falling through the focal point of the optical probe which throws off the size measurements that are collected. In the current experiments the particles that we are fed into the reactor are sieved to a narrow size bin, diminishing the importance of this measurement.

Results: In this experimental campaign we varied the reaction conditions for the combustion of two different ranks of coal char: Black Thunder, and Pittsburgh. The ranking for coal Black thunder is a sub-bituminous coal, and Pittsburgh is bituminous. We varied the environment by carrying out the reaction with either CO_2 or N_2 as the diluent gas, with 12, 24, and 36 volume % oxygen concentration, at 50, 80, 100, and 200 standard liters per minute of gas flowing through the reactor, and reactor/gas temperatures of 1200, 1400 K, at pressures slightly above atmospheric. The collected trends are shown in the Graph 1.1.



Graph 1.1: Char Comparison over Various Parameters

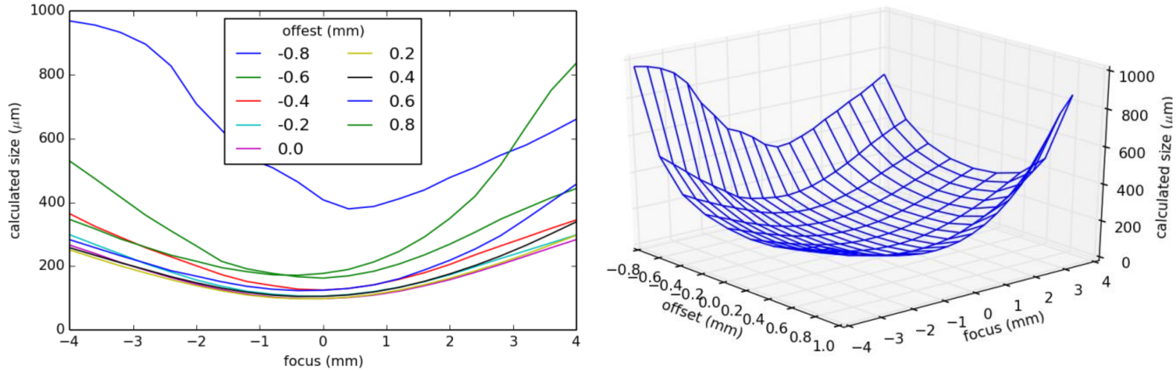
Looking at the data we see that residence time, which is dependent on the flow rate, does not change the combustion temperature significantly, causing temperature variations for each combustion environment of less than 50 K. However, for the Pittsburgh char we did have to move the optical probe down one port on the reactor to collect data since the particles were reacting later than the more reactive Black Thunder char.

Particle temperatures do increase with increasing reactor wall/gas temperature, although particle temperatures do not increase nearly as much as the wall/gas temperatures (200 K). When the volume-% of O_2 (based on total volume of gas flowing through the reactor) is increased we see much higher particle temperatures. The reason for this is because at higher oxygen concentrations there is more O_2 diffusion through the boundary layer of the fuel and it increases the oxidation rate of the combustion reaction [1].

Particle surface temperatures are significantly lower in a CO_2 environment for two reasons. First, the diffusivity of O_2 in N_2 is higher than that of O_2 in CO_2. This allows more oxygen to reach with the char particle with a nitrogen diluent [1]. Secondly, the CO_2 can directly react with the carbon in the coal, forming CO. This reaction is endothermic, which will cool the particle surface temperature [2].

When we compare Black Thunder and Pittsburgh char in a CO_2 rich environment we see interesting results due to the gasification reaction associated with coal and carbon dioxide. Black Thunder is a lower rank coal, which means that it's has less structured carbon bonds and higher reactivity than Pittsburgh coal which is one grade higher. Because of this we expect to see the more reactive coal to have higher surface temperatures during combustion, and while this is true for the N_2 diluent we don't see the same results for the CO_2 environment, for 24% O_2 (or 76% CO_2). Because Black Thunder is more reactive it goes through more gasification reactions, where the carbon dioxide reacts with the coal instead of just the oxygen with the following reaction: $C_{(s)} + CO_2 \rightarrow 2CO$. The trend of these results is also seen in a similar experiment carried out by Shaddix et al. [4], in a smaller scale furnace where the gasification reactions of higher grade char compared to a lower grade give less variance in particle temperatures for high concentrations of CO_2**.**

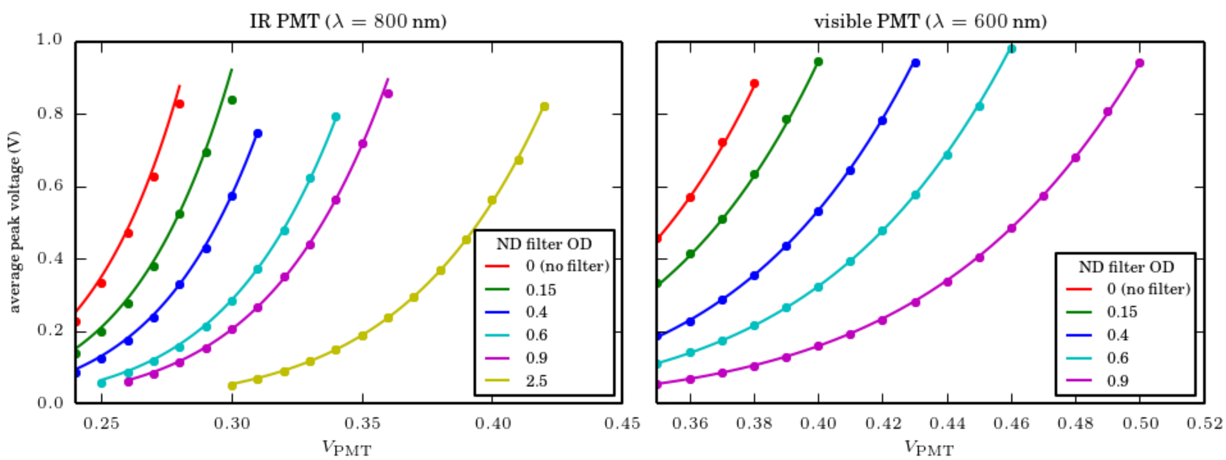
Calibrations and Error Analysis: We began error analysis for our system detection of particle size as well as continuing on the calibration constant for our temperature readings. We use a tungsten filament light source for these experiments, shown in Figure 1.3, for these calibrations. A 100 μm pinhole is attached onto a chopper wheel, with a lamp mounted behind it. The lamp is turned on, and the wheel rotates, providing moving light that is captured by the probe and the detections systems. The lamp operates at a known temperature, and the pinhole limits the simulated "particle" size. Size error analysis was done by seeing how much offsetting the distance between the collection optics and the chopper wheel, as well as the transverse location of the optics affected the size measurement. We started at a point that gave initially gave a minimum size calculation, and called that the origin point. The size measurements are graphed as a function of the offset and focus distance from the origin, shown in Graph 2.1.



Graph 2.1: Focus-Offset Adjustment for Particle Size Error Analysis

The data shows the large variation is size if a particle is out of focus, showing the need to implement a system that can detect when the particles are in focus.

Next, we looked at corrections for attenuation by the neutral density (ND) filters and the photomultiplier tube (PMT) gain to yield a system response constant. The simulated 'particle' response curve is fit to the voltage response giving a maximum signal. That signal is then graphed as a function of the filters, which is wavelength dependent and the PMT control voltage, which is logarithmically dependent on circuit voltage. The data is shown below in Graph 2.2:



Graph 2.2: PMT response to different voltages and neutral density filters, with fits shown as lines

Using this data and programming it into the LabView data acquisition allows for more accurate temperature readings with different combinations of ND filters and PMT voltage.

<u>Future work:</u>

We plan to complete data set for Pittsburgh char at current pressures, as well as looking at subbituminous char and other rank coal chars at pressures up to 10 bar and higher, currently we have done the experimentation up to 5 bar. The temperatures of these particles will be measured as a function of total and partial oxygen pressure.

A further analysis of the data is required to quantify the kinetics parameters and create char consumption models that fit our measured data. More development is planned to improve the focal point discrimination for each particle, either by improving the data analysis routines through the LabView programs, or by adding in another probe (also inserted into the reactor perpendicularly) with a laser trigger that follows the design of a smaller scale system.

Impact on Laboratory and National Missions:

This work brings a deeper understanding of combustion reactions in different environments allowing us to analyze and model trends for a wide variety of situations, and make accurate predictions to use oxy-fuel combustion in industry settings. The kinetic models can be used in CFD codes to design and optimize pressurized oxy-combustion power plants with CCS. This will allow the U.S. to continue to generate power from our vast coal reserves while reducing the environmental impact of CO_2, and still being viable and cost-effective in the current economy.

Conclusions:

The optical probe and cold-tip work well in oxy-combustion environment, and can measure temperatures of individual particles going through the reactor. The data is consistent with other studies carried out in similar experiments. When we increase the O_2 concentration we see a large increase in particle temperature and higher reaction temperatures in N_2 diluent when compared to CO_2 diluent. The flow rate/residence time and the furnace wall temperature do not greatly affect the reacting particle temperature. When comparing the two different ranks of coal, we see the effects of CO_2 gasification pulling down the particle temperatures is more significant for the lower ranked coal char. Black Thunder has higher reactivity than that of Pittsburgh, but its combustion temperatures are close to or slightly lower than Pittsburgh in a CO_2 rich environment. We’ve made headway in particle size error analysis through the calibration experiments showing how much the data can vary if the falling particles are just slightly out of the focal point of the probe optics. Further calibration of the detection systems as a function of PMT voltage and ND filters lets us determine more accurate temperature readings for a quantitative analysis.

Acknowledgements:

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[3] D.A Tichenor, RE Mitchell, KR Hencken, S Niksa: *Simultaneous In Situ Measurement of the Size, Temperature and Velocity of Particles in a Combustion Environment*, Twentieth Symposium (International) on Combustion/The Combustion Institute, 1984/pp. 1213-1221

[4] CR Shaddix, TC Williams, S Jimenez, SM Lee: *Influence of Coal Rank and Gas Temperature on Oxy-Fuel Combustion Properties* ,Combustion Research Facility Sandia National Laboratories

Appendix:

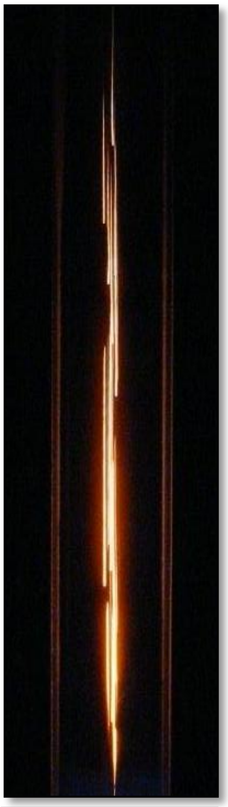


Figure 1.1: Example coal char particle light emissions

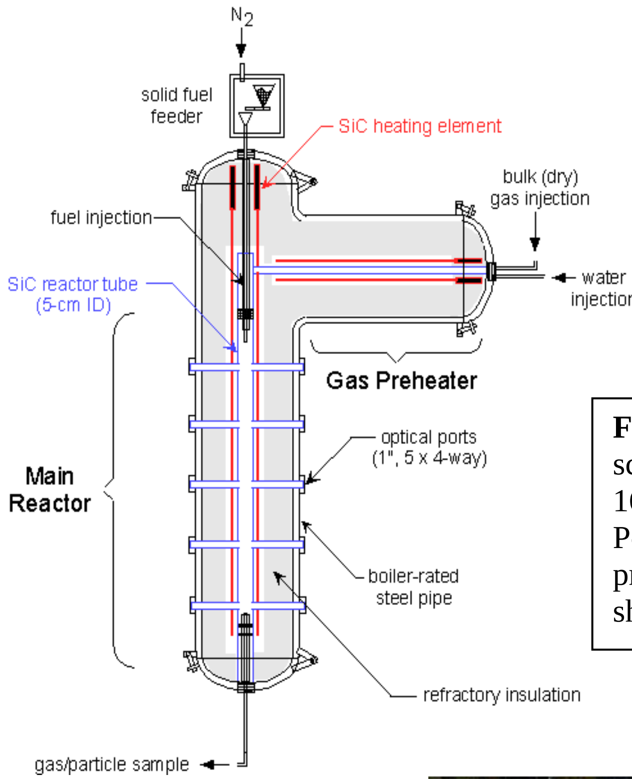
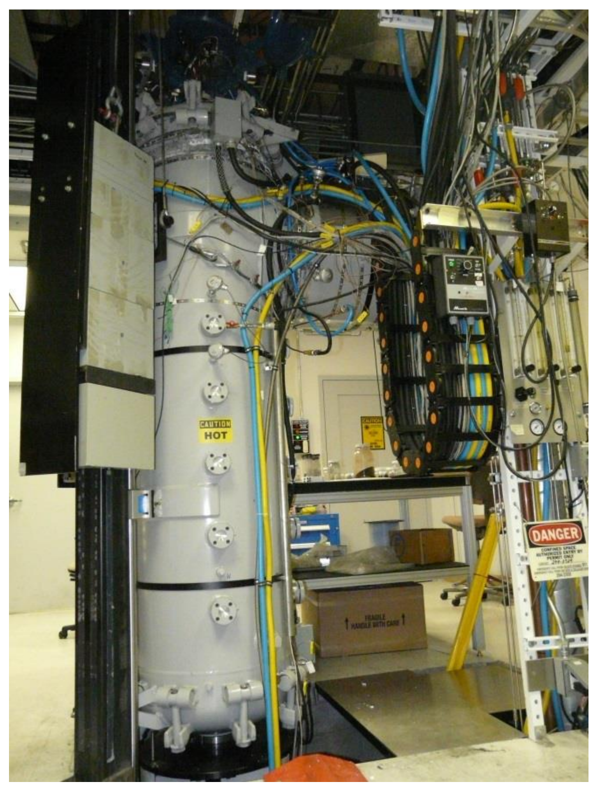


Figure 1.2: PCGR and schematic. Capable of 1600 ºC at 300 psi. Ports for the optical probe, and cold tip are shown.



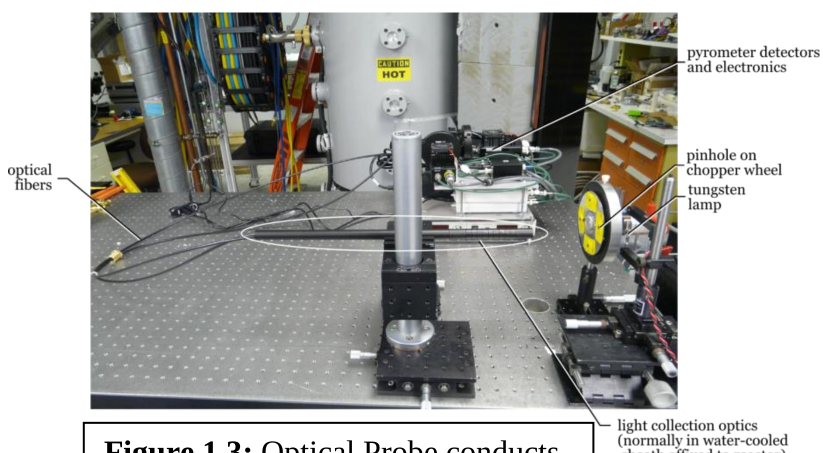


Figure 1.3: Optical Probe conducts collected light to a two color pyrometer via a fiber optic bundle

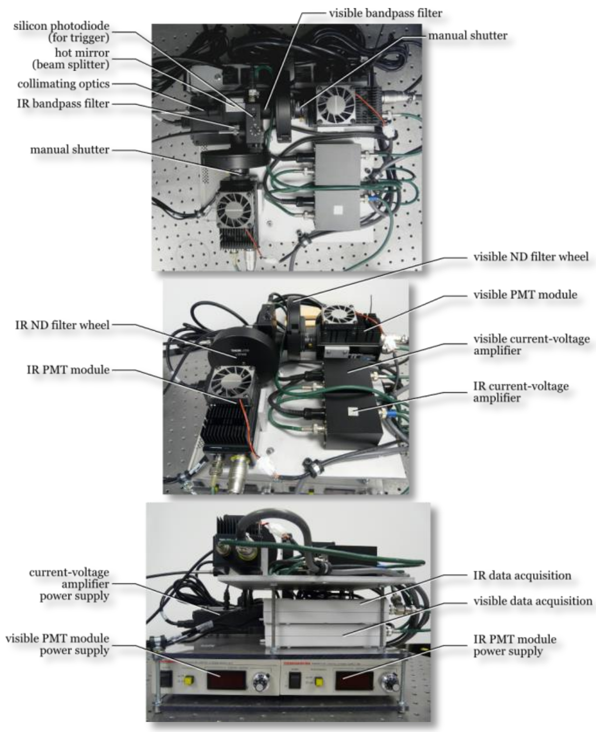


Figure 1.4: Pyrometer optics, electronics and data acquisition

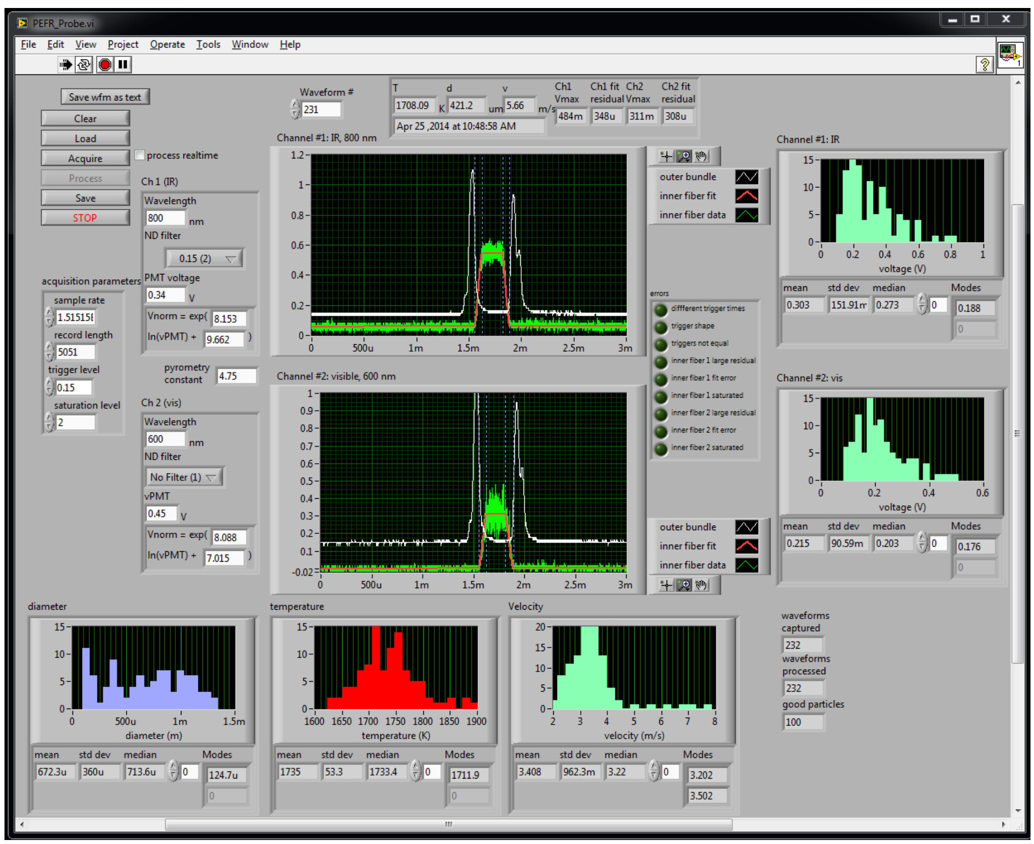


Figure 1.5: Labview data acquisition and analysis interface.